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The impact of climate change on vadose zone pore waters and its implication for long-term monitoring

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Abstract

Protecting groundwater is of growing interest as pressure on these resources grows. Recharge of groundwater takes place through the vadose zone, where complex interactions between thermal-hydrological-geochemical processes affect water quality. Monitoring processes in the vadose zone is an important means of evaluating the long-term health of aquifer systems, and has become an integral part of many subsurface engineering efforts. Monitoring such systems, however, may be affected by changes in climate that slowly propagate through vadose zone systems. We describe in this paper the use of NUFT-C, a reactive transport simulator designed to run on a high performance, massively parallel computer, to compare quantitatively the evolution of a deep vadose zone with changes expected from an engineered high-level nuclear waste repository. The results suggest that the impacts from waste emplacement are, in some instances, similar to those that would be observed as a result of climate change, whereas others are distinguishable from evolution of the natural system. Such simulations facilitate design of long-term monitoring programs that take account of these complex effects. The results emphasize the importance of developing long-term baseline measurements and control sites, in order to enhance confidence in interpretations of complexly evolving data sets that will be obtained from multi-decade monitoring efforts.

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Keywords: Vadose zone; Pore water chemistry; Climate change; Monitoring; Reactive transport; Nuclear waste disposal; Massively parallel computers

1. Introduction

The vadose zone is that region between the soil zone and groundwater aquifers through which recharge of the aquifers takes place. Scientific interest in the vadose zone has grown over the last 30 years as a result of increasing appreciation for the complexity of interactions in this environment that influence groundwater quality (Bosma et al., 1993; Tompson, 1993; Wang et al., 1997; Looney and Falta, 2000). Ultimately, this interest has contributed to a growing realization that protecting groundwater quality requires, among other things,

sophisticated long-term monitoring programs in the vadose zone (Carrigan, 2000; Looney and Falta, 2000).

Protecting groundwater quality is a pressing need. Growing human populations are accelerating pressures on shrinking groundwater resources. It is estimated that over 50% of the US population depends on groundwater aquifers for their water supply (Solley et al., 1993), while simultaneously, extraction of water from aquifers is occurring at rates greater than the recharge rate in some regions (Alley et al., 1999). The extent of this problem is expected to grow as populations increase. Similar patterns of “water-stress” are developing globally. The current rate of groundwater depletion is approximately $1.6 \times 10^{11} \text{ m}^3/\text{year}$ (Brown, 2001). If depletion continues the number of people on the planet

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living in water-stressed countries will increase from 500 million to 3 billion over the next 25 years (Postel, 2001).

Monitoring programs intended to last decades to hundreds of years are now mandated by regulation for some engineered or remediated subsurface vadose zone sites. For example, regulations stipulate that a long-term monitoring program (called the “performance confirmation period”) be developed to assure that the potential high-level nuclear waste repository proposed at Yucca Mountain, Nevada, evolves in a manner consistent with the safety case upon which the repository operating license would be granted (Lindner, 2000). This Performance Confirmation Period is required to last until closure of the repository, which may not occur for > 100 years. The proposed repository would be excavated at a depth of ca. 300 m, which is more than 200 m above the static water table. The purpose of this safety case, ultimately, is to assure preservation of the quality of the groundwater resource in this region. Similar requirements have been developed for the long-term stewardship of many sites that have been designated “Super Fund” sites, and for which site clean-up is in progress. Designing, implementing and maintaining such long-term monitoring efforts in the vadose zone poses unique scientific challenges (Roadmap, 2001; Looney and Falta, 2000), including identifying what parameters and properties can be measured, where to measure them and how to interpret the resulting data streams.

Relatively recently, independent research focussed on the vadose zone as a potential repository of climate change data (Foster and Smith-Carrigan, 1980; Allison et al., 1985; Sharma and Hughes, 1985; Barnes and Allison, 1988; Allison et al., 1994; Tyler et al., 1996; National Research Council, 2000), has implicitly raised an issue for these long-term monitoring efforts that has not yet been thoroughly considered. Reconstruction of climate records from observable subsurface properties, e.g., pore water composition (Walker et al., 1991; Campbell et al., 1996), mineralogy (Paces et al., 1996) and temperature (Lachenbruch et al., 1982) document that the vadose zone is a dynamic environment within which pore water chemistry, gas chemistry, and temperature are in a responsive state, driven by climate change at the surface. Monitoring programs in deep vadose zone settings similar to those in the US southwest, where travel times of infiltration are measured in hundreds to thousands of years, must have supporting models able to distinguish between climate-induced changes and those resulting from human activity. This distinction must be made with sufficient precision and accuracy to establish with reasonable certainty that changes observed through a monitoring program are consistent with the predicted performance of the monitored-engineered structure.

It is only within the last few years that scientific and computational capabilities have evolved sufficiently to

support the development of the needed quantitative models. Minimum requirements for these models is the ability to conduct 2D and 3D numerical simulations of coupled thermal-hydrological-geochemical (THC) interactions in non-isothermal, kinetically dominated, multi-phase systems in unsaturated porous geological media (Evans and Nicholson, 1986; Looney and Falta, 2000). Although the ability to conduct such simulations has dramatically improved (Wang et al., 1997; Viswanathan et al., 1998; Simunek et al., 1999; Glassley et al., 2001; Sonnenthal and Spycher, 2001) and available computational power has increased with the advent of massively parallel computers, recent discussions have raised important questions about uncertainty and the predictability of such systems, at least from the hydrological perspective (Entekhabi, 2001). Thorough evaluation of this question, as it relates to coupling geochemistry to thermal-hydrology, has not yet been accomplished.

In this report we compare the effects, in a typical vadose zone system, of local climate-induced changes to those changes induced by emplacement of high-level nuclear waste in a geological repository. The simulations were conducted using NUFT-C, a new THC reactive transport code that was specifically designed to run on a massively parallel IBM-SP 2, 1200 processor high performance platform (Glassley et al., 2001). The results provide baseline information about the dynamic range of chemical and mineralogical properties that might be observed as a vadose zone system naturally evolves. Such a baseline is necessary in order to recognize the signal that might be produced in response to human engineering activities. The results demonstrate that effects associated with climate change can have a large impact on pore water chemistry, and may confuse interpretation of coupled THC effects resulting from human activities, if adequate baseline information has not been collected prior to initiation of engineering activities.

2. Description of NUFT-C

NUFT-C (Nitao, 1998; Glassley et al., 2001) simulates multi-component, multi-phase, saturated and unsaturated reactive transport for non-equilibrium, non-isothermal porous media. The thermal-hydrological properties of the code have been detailed previously (Nitao, 1998) and will not be discussed here. Aqueous speciation reactions are computed on the basis of thermodynamic data supplied by the user. Activity coefficients for the aqueous species are computed using an extended Debye–Huckel method. It is assumed that aqueous speciation reactions achieve instantaneous thermodynamic equilibrium. Dissolved gas species are also included, accounting for equilibrium thermodynamic exchange between liquid and coexisting gases. An

unlimited number of aqueous species can be considered. The thermodynamic database used for computing mass action relationships can be from any source. The databases we have employed are those currently supported within the GEMBOCHS package (Johnson and Lundeen, 1994).

Dissolution of existing minerals, or precipitation of saturated and supersaturated mineral species are modeled using kinetic equations of the transition state theory form

$$R = S_A k T_{\text{fac}} \alpha \Phi \Pi a_i (1 - Q/K_{\text{eq}})^\omega,$$

where R is the reaction rate ($\text{mol/m}^3 \text{s}$), S_A is mineral surface area per cubic meter of porous medium, k is the far-from-equilibrium rate constant ($\text{mol/m}^2 \text{s}$), T_{fac} is an Arrhenius temperature correction factor, α is a power function dependent on Q/K_{eq} that accounts for changes in reaction rate close to equilibrium, Φ is a user specified factor that allows precipitation rates to be specified as a function of the dissolution rate, a_i is a factor allowing rate dependence on the activities of specified components, Q is the activity product of the components in the heterogeneous reaction, and K_{eq} is the temperature-dependent equilibrium constant. ω is a power dependence determined on the basis of empirical data that relates to the dissolution/precipitation mechanism.

Surface areas of mineral phases that are initially present are specified for each rock unit in the simulation. Surface areas evolve during dissolution or precipitation according to user-specified mineral-specific surface-area-to-volume laws. Temperature-dependent equilibrium constant values come from Johnson and Lundeen (1994).

The volume of each mineral phase dissolved or precipitated at each time step is used to recompute the porosity in each cell of the computational mesh. This change in porosity is the basis for establishing the time varying permeability evolution of the rock units. The functional relationship between porosity and permeability is treated as Kozeny-type relationship

$$K/K_0 = (\phi/\phi_0)^3,$$

where K and K_0 are the permeability and initial permeability, respectively, and ϕ and ϕ_0 are the porosity and initial porosity, respectively. Other functional relationships between porosity and permeability can be readily implemented in the code.

The structure of the code allows an unlimited number of discrete chemical and mineralogical initial and boundary conditions to be represented, thus allowing any configuration of mineralogically or chemically distinct systems to be portrayed. Time-varying chemical or thermal-hydrological inputs are also accommodated.

The physical framework through which flow occurs is represented as a continuum or a series of continua. For fractured porous media, such as volcanic rocks, use of a dual continuum model allows the mineralogies and pore

water and gas chemistries of fractures and matrix to be treated separately. Since interaction between these continua is also considered, fracture-matrix thermal-hydrological and chemical/mineralogical interaction can be represented and monitored. This methodology also allows evaluation of the consequences of fracture and matrix transport on the mineralogical and chemical changes occurring along a flow pathway. If additional continua are needed, the code can utilize an unlimited number of them in a simulation.

The Message Passing Interface standard protocols were implemented to facilitate parallel computations (MPI, 1995; MPI-2, 1997). This standard allows maximum efficiency to be achieved for parallel computations.

Internal mesh generation capabilities are built into NUFT-C. These capabilities allow global definition of cell attributes (chemical, mineralogical and physical properties, for example) in the input file, thus simplifying the initial descriptions of the data structure.

Numerical solution of the large sets of partial differential equations inherent in these calculations has been accomplished by use of the Portable, Extensible Toolkit for Scientific Computation (PETSc) (Balay et al., 1997). Although NUFT-C can use other solver packages, PETSc was chosen for this application because of its extensive library of routines, and its high level of support on the parallel machines used in these calculations.

A suite of routines was developed to allow visualization of both performance and simulation results while a simulation is running as well as after simulations are complete. These capabilities include graphics packages showing changes in specific solution components and mineral abundances, changes in temperature, saturation, gas fraction and pressure, time derivatives of the abundance of specified chemical components and minerals, changes in the magnitude of time steps as a function of number of time steps, and the simulated time versus the elapsed time.

Validation of the code was accomplished by comparing simulation results with those of other benchmark codes {EQ3/6 (Wolery, 1992a,b); GIMRT (Steefel and Yabusaki, 1996; Steefel, 2000)} for constant temperature, fully saturated systems. Comparison has also been made with bench scale experiments, field scale experiments, and field studies (Glassley et al., 2001).

3. NUFT-C performance

A typical coupled THC calculation done using NUFT-C will consist of local and global computational phases with different performance characteristics.

The local computational phase consists of calculations involving only the variables in each grid cell.

Parallelization of these local calculations is straightforward and the performance of this phase is high. The calculations performed at each grid cell are identical, except for the number of variable switching iterations in the thermal-hydrology and chemistry models, so static load balancing by assigning an equal number of grid cells to each processor is generally effective.

The global computational phases consist primarily of the linear solver portion of the solution for the partial differential equations. This is the only phase involving extensive interprocessor communication. We used the parallel linear solver from the PETSc software package, modified by us to address specific needs arising from this coupled calculation.

The linear system which must be solved globally in the thermal-hydrology model is the most difficult calculation numerically. The system is not symmetric, not positive definite and very poorly conditioned. In a typical calculation in which heat is added to the system and the temperature changes, the thermal-hydrology conditions become more complex as multi-phase flow, boiling, condensation and widely different flow velocities develop in different regions of the computational domain. As a result the linear systems become more and more stiff. Through experimentation with the GMRES (and other) solvers with Additive Schwartz Method preconditioning, a two-level multi-grid preconditioner, and a Multiplicative Schwartz Method preconditioner, we established that the latter, with the GMRES linear solver, provides the best performance for this application. Even so, as the system becomes stiffer, time stepping is often reduced to seconds or minutes in a complex calculation.

Although the linear system in the thermal-hydrology model is the most difficult calculation, in a typical moderate size problem with complex chemistry, thermal-hydrology only accounts for a small percentage (<10%) of the run time. This results from the fact that the chemistry model usually requires the use of smaller time steps than the thermal-hydrology model, often by a factor of 10 or more. Also, the local calculation phase of the chemistry model is far more complex than any other calculation phase and represents the bulk of the run time. There is (at least) one global linear system solution performed at each chemistry time step. These global systems are larger than the thermal-hydrology linear systems, but are far simpler numerically. It is not unusual for these linear systems to converge in one or two iterations using the simplest PETSc solvers and they represent an insignificant part of the run time.

4. Simulation model and approach

The purpose of this paper is to compare the effects on vadose zone pore water chemistry of climate change and

thermal perturbation from a nuclear waste repository. Details of those respective simulations are presented elsewhere (Glassley et al., 2002a, b). Here, we summarize the relevant results and develop the comparison.

The geological framework used in these simulations consisted of the thermal-hydrological units at Yucca Mountain, Nevada. Mineral abundances and surface areas for each rock unit were those described in Yucca Mountain Project reports (Sonnenenthal and Spycher, 2001), with the addition of an overburden of alluvium that was assumed to act as the interface between the volcanic stratigraphy and the atmosphere. The properties of the alluvium were those of a coarse sand, with a mineralogy identical to that of the immediately underlying rock unit.

A dual porosity-dual permeability continuum (DKM) model was used to monitor fracture and matrix mineral and chemical evolution in each of the 17 lithologic units. Although DKM models have intrinsic limitations in their ability to represent mineralogical and chemical heterogeneity and fracture-matrix interactions (Pruess et al., 1999; Glassley et al., 2002c), the DKM approach was employed in this study because it corresponds to that generally used for TH simulations in the Yucca Mountain Project.

Thirty-four minerals and 58 aqueous species (Table 1) described by the chemical components $\text{CaO-Na}_2\text{O-K}_2\text{O-Al}_2\text{O}_3\text{-MgO-SiO}_2\text{-CO}_2\text{-SO}_4\text{-H}_2\text{O}$ were used in this study. A variety of initial pore water compositions were considered, all restricted to waters that would be classified as chloride-bearing calcium or sodium bicarbonate waters (Garrels and MacKenzie, 1967; Eugster and Hardie, 1978; Rosenberg et al., 2001). Initial pore gas was assumed to be 300 ppmv CO_2 , but was allowed to evolve during initialization (see later). This resulted in a CO_2 fugacity that was consistently below ambient atmospheric values. These results provide constraints on the system behavior for conditions in which CO_2 fugacity is low, relative to that in the current atmosphere, thus complementing other simulations conducted at higher CO_2 partial pressures (e.g., Sonnenenthal and Spycher, 2001). In this report we discuss the key results from a simulation in which it was assumed that the initial pore water was that reported in Sonnenenthal and Spycher (2001), and the surface influx was assumed to have the composition of local rainwater (Yucca Mountain Project, 2000). It is likely chemical modification of the infiltrating water occurs within a soil zone in the natural system, which we do not consider here. Future studies are planned that evaluate this effect.

The protocol followed for the climate change simulations was to conduct an initialization run, allowing the system to achieve steady-state conditions for saturation, temperature, pressure, and liquid and gas chemistries in all lithologic units. After steady-state conditions persisted for more than 20,000 years, the system was

Table 1
Primary and secondary mineral and aqueous species used in simulations

Primary aqueous species	Secondary species ^a		Primary minerals	Secondary minerals
H ⁺	OH ⁻	HCl	Albite	Analcime
Ca ⁺⁺	AlO ₂ ⁻	KOH	Anorthite	Boehmite
K ⁺	AlO ⁺	KHSO ₄	Calcite	Chalcedony
Na ⁺	AlOH ⁺⁺	KSO ₄ ⁻	Clinoptilolite-Ca	Clinochlore
Mg ⁺⁺	HalO ₂	MgHCO ₃ ⁺	Clinoptilolite-Na	Dolomite
SiO ₂	CaHSiO ₃ ⁺	MgCl ⁺	Cristobalite	Gibbsite
Al ⁺⁺⁺	CaOH ⁺	MgOH ⁺	Glass	Gypsum
HCO ₃ ⁻	CaCl ⁺	MgHSiO ₃ ⁺	K-Feldspar	Halite
Cl ⁻	CaCl ₂	MgCO ₃	Montmorillonite	Heulandite-Ca
SO ₄ ⁻	CaHCO ₃ ⁺	MgSO ₄	Quartz	Heulandite-Na
O ₂ (gas)	CaHSiO ₃ ⁺	NaCl	Amorphous SiO ₂	Hydromagnesite
Tracer ^b	CaCO ₃	NaOH		Kaolinite
	CaSO ₄	NaHSiO ₃		Laumontite
	ClO ⁻	NaSO ₄ ⁻		Mordenite-Ca
	ClO ₂ ⁻	O ₂		Mordenite-Na
	ClO ₃ ⁻	HSiO ₃ ⁻		Natrolite
	ClO ₄ ⁻	HSO ₄ ⁻		Paragonite
	HClO	HS ⁻		Pyrophyllite
	HClO ₂	S ₂ ⁻		Scolecite
	CO ₃ ⁻	S ₂ O ₃ ⁻		Sepiolite
	CO ₂	SO ₃ ⁻		Sylvite
	CO ₂ (gas)	H ₂ S		Talc
	KCl	SO ₂		Wairakite

^a Dissolved secondary aqueous species, unless indicated as (gas), in which case the species was included in the gas phase.

^b Fictive, non-reacting aqueous component used to track fluid movement.

perturbed in a manner that approximates the magnitude and time duration of change between past pluvial conditions and the present climate. For this simulation the infiltration flux was changed from 87.5 to 8.75 mm/year, and temperature changed from 12.7°C to 17.7°C over a 10,000 year period. We selected these conditions on the assumption that they would be illustrative of the character of change that may have occurred over that time period (Spaulding, 1985). However, we recognize that the precise conditions 10,000 years ago are unknown and changes in conditions would not have followed a smooth evolution. For this reason we emphasize that the results presented here should only be viewed as illustrative of the system dynamics to be expected in deep vadose zones.

For those simulations in which the geological framework was subjected to thermal perturbation by emplacement of high-level nuclear waste, the system was modeled in 3D assuming that 60 waste packages were emplaced in two tunnels separated by ca. 80 m. The waste packages were treated as individual heat sources, each with its own time-dependent thermal output. The computational mesh, which extended over 2 km² of ground surface and 550 m depth, consisted of approximately 10⁶ cells that varied from ca. 10 cm on a side near the waste emplacement tunnels, to ca. 10 m on a side a kilometer from the tunnels. This spatial configuration

approximately represents an outer corner in a nominal Yucca Mountain-like repository design.

We present the results of the effects of waste emplacement for the last 15 years of a 100 year Performance Confirmation Program. We focus attention on this period because it provides a clear example of the issues that must be considered when designing and implementing such a monitoring effort.

Comparison of these two suites of simulations allows identification of regions where a subsurface monitoring program could expect to obtain unambiguous indications of the response of the mountain to waste emplacement, and where the results may be ambiguous because the rate of change of variables for the naturally evolving system is similar to that for the perturbed system.

5. Results

The ambient thermal conditions are a combined result of heat transfer from the surface and advective removal of heat to deeper levels via infiltrating water. Thus, a higher surface temperature will tend to increase the ambient temperature at depth, as will a lower infiltration flux. For the case in which we consider the change from pluvial to present conditions, the temperature increased

by approximately 8°C at 200 m, and increased by approximately 9°C at 400 m. This change results in a steeper vertical temperature gradient (0.0159°C/m after perturbation vs. 0.0122°C/m before perturbation) over this depth interval. The maximum rate of temperature change is $\sim 0.0009^\circ\text{C}/\text{year}$, but is depth dependent (Fig. 1).

At the new infiltration flux of 8.75 mm/year, complete replacement of in situ fracture water is accomplished within ~ 2000 years at both depth intervals (~ 200 and ~ 400 m). The largest fluid reservoirs in the system, however, are the matrix blocks that account for more than 90% of the total fluid volume. Exchange of water between the matrix and fracture continua is primarily through capillary imbibition. As a result, complete pore water replacement takes much longer in the matrix, requiring more than 13,000 years at ~ 200 m and 21,000 years at ~ 400 m.

Controls on pore water chemistry are primarily changes in the ambient temperature, and the residence times and volumes of solutions in fracture and matrix pores. The matrix water is primarily affected by the increase in temperature, since matrix saturations are always very high. Fractures, on the other hand, experience complex changes in saturation as well as an increase in temperature, resulting in more complex evolution. The rate at which chemical change occurs is dependent upon the exposed surface areas of the solid phases in contact with the solution, as well as the degree of mineral supersaturation or undersaturation. Given

that the dissolution and precipitation rate constants for the minerals in this study range between 10^{-7} and 10^{-14} mol/cm²/s, and that initial mineral abundances and surface areas in each lithologic unit are different, pore water evolution differs between and within lithologic units. The time required for solution composition to recover to a steady state will, therefore, be no shorter than the time required to achieve steady-state temperature and liquid saturation conditions.

The combined temperature and saturation effects lead to complex evolutionary pathways for the fracture and matrix pore water chemistries. There is a small decrease in the aqueous SiO₂ concentration in the fractures ($\Delta \log \sim 0.02$ – 0.06) but a relatively large increase ($\Delta \log \sim 0.25$) in the matrix (Fig. 2). The net result, at steady state, is that the aqueous SiO₂ concentration ratio between fracture and matrix pore waters changes from ~ 1.5 to 0.8.

The log of the bicarbonate concentration increases in both fractures and matrix by ~ 0.15 (Fig. 3). The ratio between fracture and matrix bicarbonate concentrations is approximately the same before and after the transition period during which surface temperature and infiltration flux are changing. However, for more than half of the transition period, the fracture and matrix concentrations reverse their relative values and the bicarbonate mole ratio between matrix and fracture change from >1.0 to <1.0 .

The results for the repository model exhibit highly localized changes that reflect the thermal impact of the

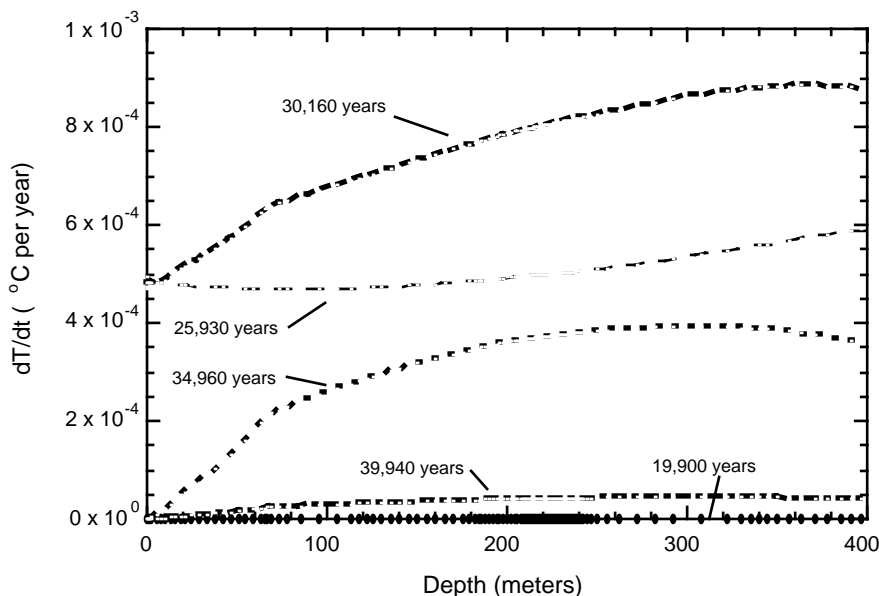


Fig. 1. Rate of temperature change as function of depth in response to changes in surface infiltration flux and surface temperature. Note that changes in infiltration flux and temperature were initiated at 20,000 years and ended at 30,000 years. Data indicated for 19,900 years documents that steady-state existed prior to perturbation of surface boundary conditions. Also, note that steady-state conditions require more than 10,000 years to be achieved after perturbation is terminated (data labeled 39,940 years).

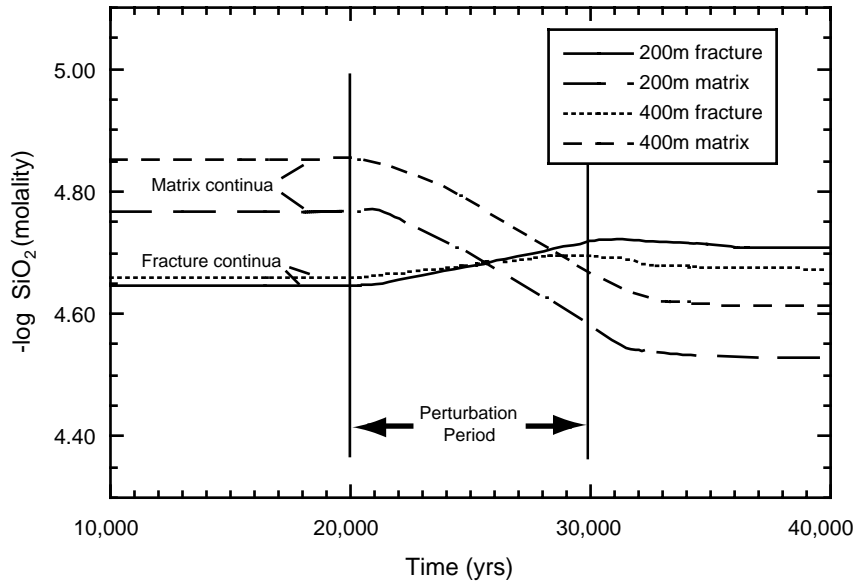


Fig. 2. Aqueous SiO_2 concentration (as negative log SiO_2 molality) in fractures and matrix as function of time at ca. 200 and 400 m, for same time period represented in Fig. 1. Shaded region indicates duration of time over which surface boundary conditions changed.

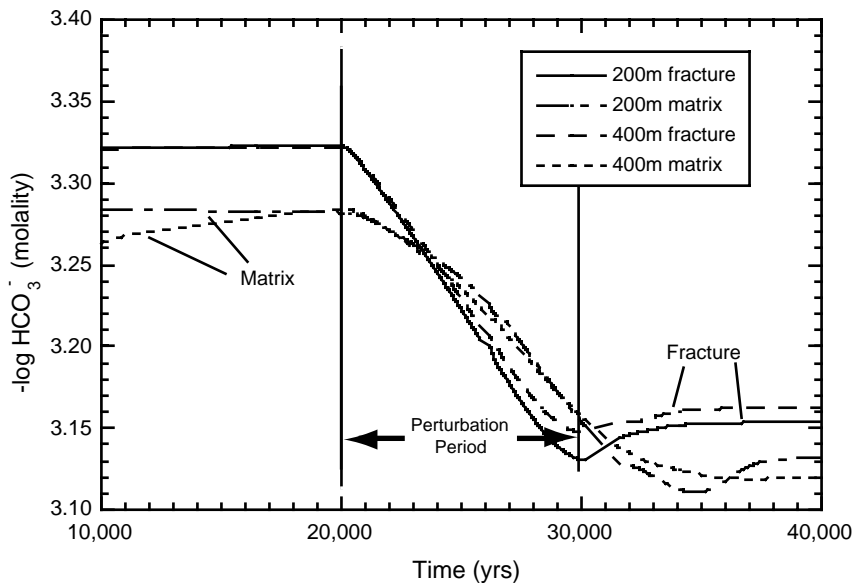


Fig. 3. Aqueous HCO_3^- concentration in fractures and matrix as function of time at ca. 200 and 400 m, for conditions represented in Fig. 1. Shaded region indicates duration of time over which surface boundary conditions changed.

waste packages. For the period of time considered, drift wall temperatures are in the high 40°C to mid- 60°C range, depending upon the heat output of the nearest waste package. Temperature isotherms are approximately concentric to the tunnel walls in cross section.

In the near-neighborhood of the drift walls, an evaporation front forms that slowly migrates outward from the

tunnels over a period of decades. The resulting water vapor condenses in cooler regions along fracture surfaces within a few tens of meters of the tunnel walls. Some of this condensate is imbibed into the porous matrix, and some migrates downward along fracture surfaces. Liquid saturation in the matrix changes at the rate of $10^{-3}/\text{year}$. The net pathway of the condensed

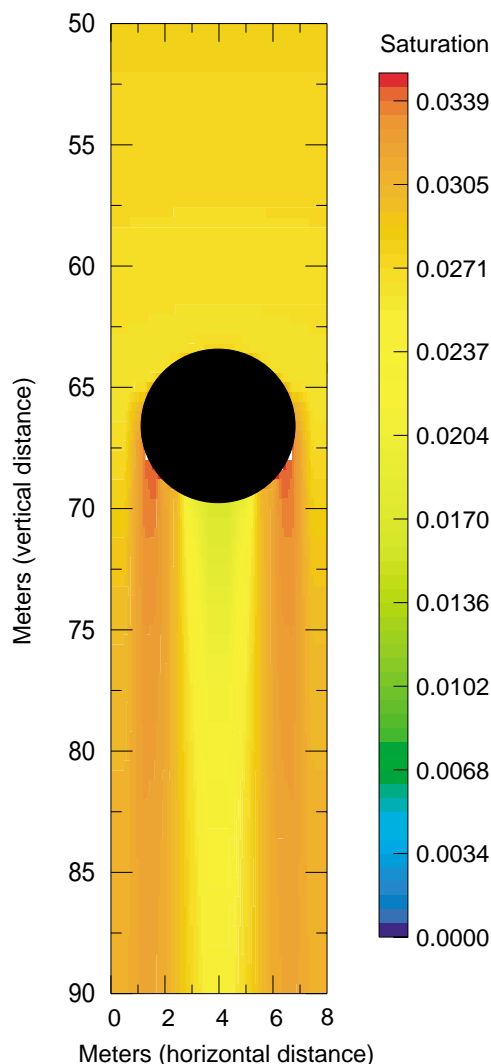


Fig. 4. Vertical cross section through one of two waste emplacement tunnels showing distribution of fracture saturation at 85 years. Vertical scale is referenced to an arbitrary zero point. Large black circle indicates tunnel.

water is around the tunnels and downward (Fig. 4). Infiltrating water, distinct from the water that condenses as a result of evaporation, is diverted around the tunnels and follows the same pathway as the condensate. This process leads to an increasingly larger volume of dried rock on either side of and above the tunnels, extending to a distance of a few meters, and formation of a “shadow” zone directly below the tunnels. These results are consistent with those previously reported for the thermal-hydrological behavior of the repository system (Buscheck et al., 1999; Buscheck et al., 2000a,b; Sonnenthal and Spycher, 2001).

Hydrological disequilibrium persists between fracture and matrix gas and liquid phases for the period of time

considered here, which is also consistent with the results of previous studies (Buscheck et al., 1999; Buscheck et al., 2000a,b; Sonnenthal and Spycher, 2001). Through the non-linear coupling between chemical and thermal-hydrological processes, chemical disequilibrium persists and steady state is never achieved.

Liquid saturation within the fractures is lowest directly below the tunnels (Fig. 4) while fractures on either side of the low saturation zone have the highest liquid saturations (~ 0.03). During the 15 year period detailed here, saturation in the fractures decreases at an annual rate of about 1 part in 10^4 . Parallel to the tunnels, fracture saturation varies by about a factor of two, reflecting the impact of the heat output of the nearest waste package.

Changes in water chemistry (detailed in Glassley et al. (2001)) are complex and differ from one aqueous species to another and from region to region. For aqueous SiO_2 , the largest changes occur directly below the emplacement tunnels, where concentrations change at a rate exceeding 1e^{-6} gm/kg of H_2O /year (Fig. 5). The remainder of the region in the vicinity of tunnels changes at rates much lower than this, and overlaps the predicted rate of change due to climate-induced effects. For HCO_3^- , regions of highest fracture saturation that extend downward from the sides of the tunnels are also the regions in which aqueous bicarbonate concentrations are the lowest. Separating these zones and located in the region of highest rate of change of aqueous SiO_2 is an area in which the aqueous HCO_3^- concentration is nearly an order of magnitude higher. Parallel to the drift, bicarbonate varies by slightly less than half a log unit, in response to variation in local temperature, which is determined by the nearest waste package. The rate of change of bicarbonate is about an order of magnitude greater than for aqueous SiO_2 , and exceeds the rate of change due to climate effects over approximately half of the region (Fig. 6).

There is very little mineralogical evolution throughout this period. The greatest mineral development is that of calcite, which develops an abundance of about 1 part in 10^5 , by volume, in some matrix regions a few meters out from the tunnel walls at about the same elevation as the tunnel floors. This is essentially undetectable.

6. Discussion

The simulations described here require specification of more than 24 thermal-hydrological variables and 10 chemistry-related variables for each rock unit, as well as a minimum of 7 variables for each mineral and chemical species. Uncertainty in those variables that concern the reaction rate, such as surface area, rate constant or activation energy, will contribute linearly to uncertainty in the time at which steady-state conditions are achieved

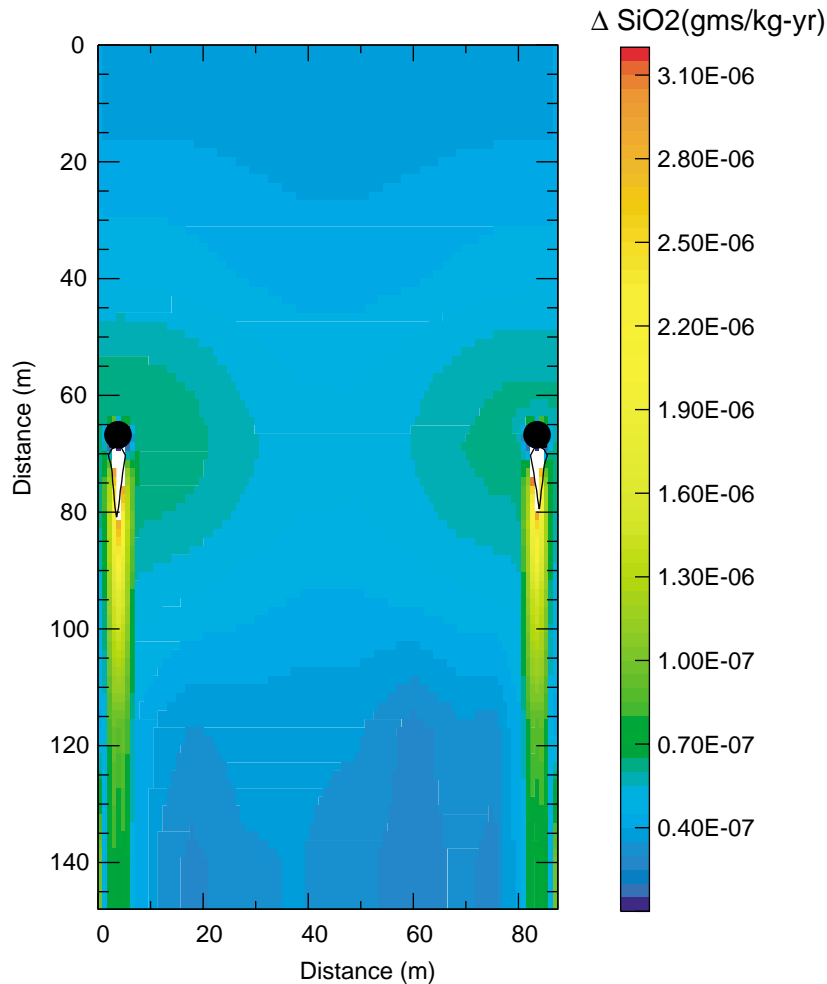


Fig. 5. Vertical cross section through two waste emplacement tunnels (centered at ~ 66 m vertical distance and 5 and 85 m horizontal distance), showing spatial distribution of rate of change of SiO_2 (g/kg of H_2O -yr) in fracture water during last 15 years of hypothetical 100-year monitoring period. Tunnels are referenced to same vertical scale as that in Fig. 4. White region maps that area in which rate of change of aqueous SiO_2 exceeds maximum rate of change computed for naturally evolving vadose zone system. Areas that have rates which overlap those of natural system are shown in color (rates are keyed to color bar).

and the rate at which they are approached. Uncertainty in thermodynamic properties of minerals and aqueous species will contribute to uncertainty in the computed aqueous and gas phase chemistries, and identification of minerals that dissolve or precipitate. Uncertainty in the thermal-hydrological properties influence the magnitude and rate of temperature changes and saturation conditions, which also effect the direction and rate of chemical reactions. It is the collection of these various uncertainties that contribute to the overall uncertainty in the computed results. However, there are currently insufficient data available to rigorously compute an uncertainty envelope for the calculations presented here (see Glassley et al., 2002a for further discussion). However, a variety of test cases, as previously

mentioned, have been used to establish that, within the constraints of the available data, the code accurately reproduces the results of experiments, field observations, and analytical solutions for a wide range of systems. In the discussion that follows, these considerations should be kept in mind.

Recent studies of climate variability in the Southwest of the United States documents significant climate fluctuations on many different time scales, some as short as ca. 100 years (Spaulding, 1985; Waters, 1989; Hostetler and Benson, 1990; Dean et al., 1996; Oviatt, 1997; Krider, 1998; Allen and Anderson, 2000). This implies that the entire vadose zone in the southwest may well be in the process of chemically and thermal-hydrologically adjusting to relatively recent, post-glacial

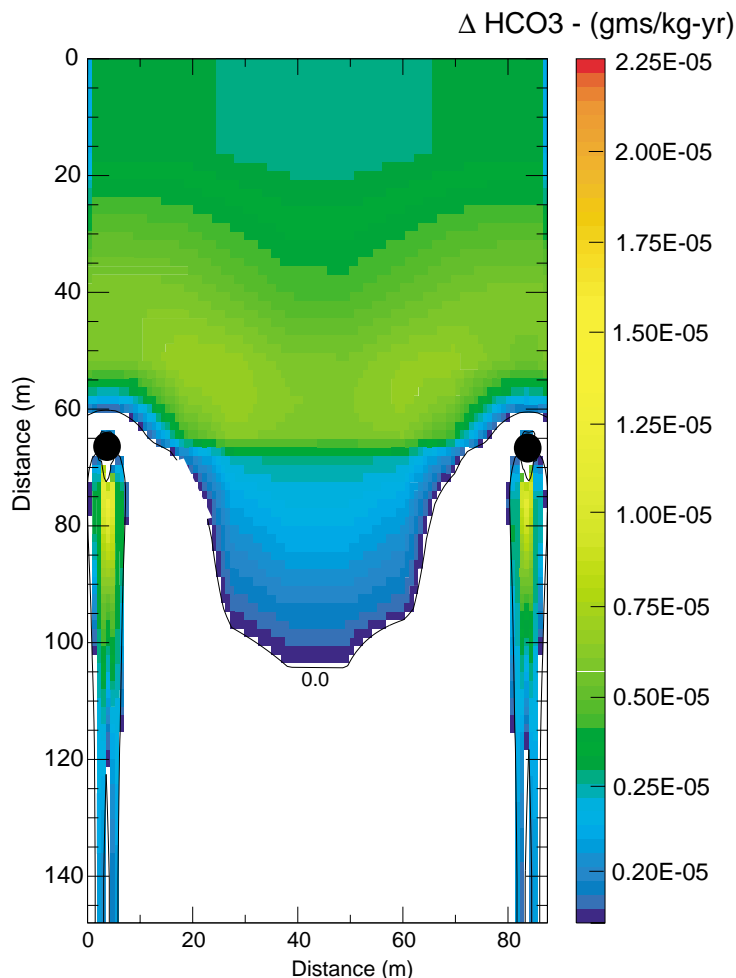


Fig. 6. Same cross section as in Fig. 5, showing spatial distribution of rate of change of aqueous HCO_3^- (g/kg of H_2O -yr) in fracture water during last 15 years of hypothetical 100-year monitoring period. See Fig. 5 caption for further explanation.

climate changes, and is not at steady-state (Phillips, 1994). The results of the simulations presented here are consistent with those conclusions, and quantify the magnitude of effects that may be expected. The characteristic response times for these systems to achieve new steady-state conditions after perturbation has ceased are on the order of thousands of years, but differ between fracture and matrix. Given the continuously evolving climate patterns over the last ca. 10^6 years or more, the present day deep vadose zone is anticipated to be in a state of flux.

Although the climate-forced rate of change of variables in the deep vadose zone is very slow, it is, in some instances, indistinguishable from the rate of change resulting from human-induced perturbations of the physical system. For example, the rate of change of climate-induced aqueous SiO_2 evolution is virtually indistinguishable from that due to effects from waste

emplacement (Fig. 5) throughout most of the region that is a likely target for monitoring. For aqueous HCO_3^- , on the other hand, the rate of change in the human-perturbed system is high enough over relatively large, well-defined volumes of rock to allow clear recognition of a signal from the waste emplacement activity (Fig. 6).

Except within a few meters of the emplacement tunnels, the rate of change of variables of interest is near or below the limit of practical measurement. However, this problem, and the issue of distinguishing between natural and perturbed signals can be overcome if a long-term monitoring program has supporting it a long-term database of measurements taken before engineering activities disturb the natural system. Such measurements can be used to establish a baseline that characterizes the current evolutionary pathway being followed by pore waters and gases in the natural system. If such measurements are of sufficient quality, then the

impact of the human-induced perturbation can be seen as changes imposed on the long-term, natural trends, even in those regions where the rate of change of natural and perturbed effects is similar. Establishing such a baseline requires sufficient monitoring time to establish the rate of change of those variables in the natural system that are amenable to measurement. This period of time will depend upon the variable of interest and maybe be a few years to tens of years. In addition, confidence that baseline trends are accurately determined and signals of engineering impacts properly identified, can be enhanced significantly by establishing monitoring sites that would act as controls throughout the Performance Confirmation Period. If carefully chosen and adequately isolated from the subsurface engineered facility, such control sites would allow recognition of variation and changing trends in the natural signal which might confuse interpretation of the data obtained at the monitoring sites within the engineered structure.

7. Conclusions

The value of a monitoring program of subsurface facilities comes from the ability to obtain data that allows testing and refinement of performance projections. The slowly evolving, dynamic character of the vadose zone poses important challenges for long-term monitoring programs. Vadose zone water chemistries are likely to be in a continuously evolving state. The simulations presented here provide an indication of the extent to which the signal from an evolving engineered facility may be hidden within the natural variability of a changing vadose zone. In the absence of a strategy to reduce uncertainties associated with variables required for simulations, and to address the complexity inherent in a naturally evolving vadose zone system, data collected from such a monitoring program may be difficult or impossible to use for testing and refining performance projections.

This difficulty can be overcome by establishing a long-term baseline study that resolves the natural evolutionary trends of the unperturbed system. To do this would require establishing an array of monitoring sites, some of which would be isolated from any engineering effects, and others that would be designed to see a signal from the subsurface facility. Inter-site comparison would allow direct and unambiguous recognition of the impact on vadose zone gas and water chemistries, provided the baseline measurement is of sufficient quality and duration.

Where to place monitoring facilities should be established on the basis of simulations similar to the ones described here, but which are more directly

constrained by both site specific data and observations, and the as-built properties of the engineered structure.

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